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J. J King

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EDF Page 1

Subject : Methodology for Determination of a Radiological Inventory for Pit 9 and Corresponding Results


Abstract:

A radiological inventory for Pit 9 of the Subsurface Disposal Area (SDA) was established by using data from the Radioactive Waste Management Information System (RWMIS), actual shipping records and documented values from the Rocky Flats Plant. Twelve radionuclides of concern are identified in the deposited waste of Pit 9. These are decay-corrected to 1991 and 1992 and include 3 additional radionuclides which are daughters of 3 of the original 12 radionuclides. Six transuranic radionuclides comprise 99.9% of the radioactivity originally emplaced in Pit 9.


Note: The calculations performed in this report have attempted to use the best (most accurate) data available. Variances with other references may reveal calculational differences in mass-to-radioactivity conversions, and vice versa, that can be as large as 10 percent.

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| Author | Dept. | Reviewed | Date | Approved | Date |
|---|---|---|---|---|---|
| J. J. King | 7830 | R. L. Nitschke | 7/10/91 | | |

# Methodology for Determination of a Radiological Inventory for Pit 9 and Corresponding Results

J. J. King
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The determination of the radiological inventory for Pit 9 must incorporate data from the Radioactive Waste Management Information System (RWMIS) for onsite shipments, shipping records, and annual radionuclide masses shipped from Rocky Flats to the INEL during 1968. The Rocky Flats radionuclide information is in a letter from Lee to Soule[1] and displays the following radionuclides and quantities for 1968:

| Radionuclide | Quantity |
|---|---|
| U-238 | 33,373 kg |
| U-235 | 1210 g |
| Pu-238 | 4.18 g |
| Pu-239 | 43,543.44 g |
| Pu-240 | 2720.83 g |
| Pu-241 | 210.11 g |
| Pu-242 | 7.44 g |
| Am-241 | 1778 g |

A review of Rocky Flats shipment records for 1968 by Rhodes[2] has revealed that 345,377 $ft^3$ of waste were sent to the INEL from Rocky Flats. Starting with October 10, 1968 shipments, various radionuclides quantities were listed with the individual shipments. Using this information, Rhodes has determined that only 203 g of Pu-239 were included in 67,352 $ft^3$ of waste sent to Pit 10 during the latter part of 1968. (No other listed radionuclides were tabulated by Rhodes.) This leaves 43,340.44 g of Pu-239 available to the remaining 278,025 $ft^3$ of waste shipped from Rocky Flats to the INEL during 1968. Rhodes has also determined that 120,103 $ft^3$ (34.8%) of Rocky Flats waste was placed in Pit 9 during 1968. If we allow the remaining Pu-239 to be distributed uniformly throughout the waste volume not associated with Pu-239 identified in Pit 10, we can use the following relationship:

(120,103/278,025) * (43,543.44 - 203)/43,543.44 * 43,543.44 g = 18,722 g of Pu-239.

If we assume this is applicable to the whole Pu family (Pu-52 using Rocky Flats nomenclature), which is reasonable for Rocky Flats waste, then the following relationship can be applied to all of the Pu isotopes:

0.431986 * 0.995338 * Mass of the ith Pu isotope = ith Pu Mass in Pit 9
or
0.42997 * Mass of the ith Pu isotope = ith Pu Mass in Pit 9

This computation is also valid if we require that the remaining Pu not identified in Pit 10 is uniformly mixed throughout the remaining waste volume. Using the above relationship, the following masses are generated for the Pu isotopes.

| Pu Isotope | Quantity(g) |
|---|---|
| 238 | 1.80 |
| 239 | 18,722 |
| 240 | 1,170 |
| 241 | 90.34 |
| 242 | 3.20 |

These revised values estimate the quantities of Pu isotopes in Pit 9.

The quantities of U-235 and U-238 are determined from the Rocky Flats masses noted above and from entries found in the waste received from onsite generators. The calculation of Rocky Flats contributions for U-235 and U-238 proceeds by multiplying the above masses for 1968 (1210 g and 33,373 kg) by the fraction of waste volume going to Pit 9 in 1968 (34.8%). This results in 419 g of U-235 and 11,600 kg of U-238. Based upon the presence of U-235, and the known mass abundance of U-234 (0.0055%) and U-235 (0.72%), an additonal 3.2 g of U-234 can be conservatively estimated to be in Pit 9.

RWMIS tallies are displayed in Garcia and Knight[3] showing 182 kg of U-235 and 514 g of U-238 associated with onsite generators. However, the first

value is actually 182 kg of natural uranium and was an entry error in RWMIS. This can be broken down to include 0.0055% U-234, 0.72% U-235, and 99.2745% U-238 by mass. This results in a breakdown of the 182 kg of natural uranium into 180.7 kg of U-238, 1310 g of U-235, and 10.0 g of U-234. The addition of the 514 g of U-238 noted above yields 181 kg of U-238 when rounded off to three significant figures.

The total mass for each uranium isotope expected in Pit 9 is below.

| U Isotope | Mass(g) |
|---|---|
| 234 | 13.2 |
| 235 | 1730 |
| 238 | 1.18E+7 |

The expected Am-241 in Pit 9 is also estimated by multiplying the fraction of waste going into Pit 9 (34.8%) times the amount of Am-241 shipped from Rocky Flats (1778 g). This produces 619 g of Am-241.

Several other categories of radionuclides are found in Pit 9. Reference 3 lists 0.301 Ci of Co-60, 0.012 Ci of MAP (Mixed Activation Products), 4.463 Ci of MFP (Mixed Fission Products), and 0.083 Ci of unidentified beta-gamma emitters. These categories were specifically denoted on shipping records and were entered into the RWMIS with these designations. These items are all generated by onsite generators.

The number and quantity of isotopes in mixed activation products depends upon the number of atoms of a target isotope present in a neutron flux, the (thermal) neutron cross-section, the half-life of the neutron-absorbing isotope and the time history of the irradiation. INEL test reactors generally have a history of intermittent operation for reasons of testing or fuel changing. The activation products produced are commonly associated with stainless steels that are components of reactor systems. Based upon analysis of constituents in 300 series stainless steel, Hartwell and Thompson[4] show that Co-60 is the predominant gamma-emmitting radionuclide for short-term reactor irradiations. This is supported by gamma-ray spectrometric measurements of Waste Experimental Reduction Facility (WERF) ash in Reference

4, which showed Co-60 and Cs-137 comprised approximately 83% of the radioactivity in a series of ash containers that were gamma scanned. The 5.271-year half-life and the 2504 keV mean gamma-ray energy per decay[5] imply that the external gamma-ray exposure due to Pit 9 MAP today and in the near future will be dominated by Co-60.

The test reactors at the INEL have been predominantly lightwater thermal reactors that have utilized enriched U-235 fuel. The determination of type and quantity of fission products in Mixed Fission Products (MFP) is dependent upon the instantaneous and cumulative fission yield (assumed to be for thermal neutrons striking U-235), the fission product half-life and the irradiation history. The half-lives of the precursors for a given fission product will also affect the buildup of that fission product, particularly if the irradiation history is periodically up and down.

A compromise of the above criteria can be determined by considering radioactive fission products that have half-lives greater than 2 years, produce a mean gamma-ray energy per decay that is notable (> 500 keV) and have a significant cumulative fission yield. Cs-137 has a cumulative fission yield of 6.1862%[6], its daughter (Ba-137m) is in equilibrium with the parent in less than 1 hour with a mean gamma-ray energy of 595 keV per decay, the precursors have half-lives of minutes or shorter[7], and Cs-137 has a half-life of 30.17 years. Ba-137m has a 2.5513 minute half-life before attaining a stable ground state.[8]

Gross measurements of gamma-ray radiation fields with short-lived fission products present (< 30 days) will overestimate the amount of Cs-137(Ba-137m) if the source is assumed to be solely Cs-137(Ba-137m). This ensures that Cs-137(Ba-137m) assignment will generally be conservative for the multi-radionuclide specific activities that may produce the radiation field at the time of measurement. The assignment of Cs-137(Ba-137m) may not predominate over longer-lived fission products for time intervals beyond three or four Cs-137 half-lives. This will depend upon the specifics of the irradiation histories for a given MFP waste for a chosen generator.

The previous paragraph has concentrated on a predominant beta-emitting fission product that is associated with a gamma-ray emmission via a shortlived (Half-life = 2.5513 minutes) daughter (Ba-137m). Another significant pure beta emitter is Sr-90 with a 28.6-year half-life and a cumulative fission yield 5.7664% (see Reference 6). It also has shortlived precursors that readily reach equilibrium. It has a daughter (Y-90) that possesses a 64.1 hour half-life before reaching a stable ground state. Y-90 is essentially a beta emitter.[9] The relatively low maximum permissible lung and bone burdens for Sr-90 in the human body makes it an important radionuclide when considering human health risk.[10]

The equations that describe the growth and decay of Sr-90 and Cs-137 during and following irradiation will lead to similar amounts of radioactivity for both isotopes due to the comparable half-lives and the shortlived precursors. The ratio of the cumulative fission yields for Sr-90 and Cs-137 is 0.932 and this will determine, to a first order, the expected radioactivity for one if the other is known. If we conservatively assume that 4.463 Ci of MFP noted above are from gamma-ray radiation fields associated with Cs-137(Ba-137m), then the amount of Sr-90 present is just 0.932 times 4.463 Ci, or 4.160 Ci.

The 0.083 Ci of unidentified beta-gamma emitters is most likely based upon smears of lose contamination of radionuclides of unknown origin. Because this number is small (and hence a negligible contribution to overall risk) for the Pit 9 inventory and because the identification of the responsible nuclides is highly uncertain, the unidentified beta-gamma emitters will not be considered as part of the working Pit 9 radionuclide inventory.

Table 1 summarizes the individual radionuclides that have been identified. The radioactivity values in Curies are shown along with the masses in grams. The relationship used for converting radioactivity in Curies to mass in grams or vice versa is given by:

Mass (grams) = 2.798E-06 * Activity (Ci) * Half-life (y) * Mass Number

As noted in Table 1, the half-lives are taken from Reference 8.

Table 1. 1968 Pit 9 Radiological Inventory

| Isotope | Activity (Ci) | Mass (g) | Half-Life (y)[a] |
|---|---|---|---|
| U-234 | 8.23E-02 | 1.32E+01 | 2.450E+05 |
| U-235 | 3.75E-03 | 1.73E+03 | 7.038E+08 |
| U-238 | 3.97E+00 | 1.18E+07 | 4.468E+09 |
| Pu-238 | 3.08E+01 | 1.80E+00 | 8.774E+01 |
| Pu-239 | 1.16E+03 | 1.87E+04 | 2.412E+04 |
| Pu-240 | 2.65E+02 | 1.17E+03 | 6.570E+03 |
| Pu-241 | 9.34E+03 | 9.03E+01 | 1.435E+01 |
| Pu-242 | 1.26E-02 | 3.20E+00 | 3.763E+05 |
| Am-241 | 2.12E+03 | 6.19E+02 | 4.322E+02 |
| Co-60 | 3.01E-01 | 2.66E-04 | 5.271E+00 |
| Co-60(MAP) | 1.20E-02 | 1.06E-05 | 5.271E+00 |
| Cs-137(MFP) | 4.46E+00 | 5.16E-02 | 3.017E+01 |
| Sr-90(MFP) | 4.16E+00 | 3.00E-02 | 2.86E+01 |

a. The half-lives are taken from J. K. Tuli, "Nuclear Wallet Cards", National Nuclear Data Center, Brookhaven National Laboratory, January 1985.

Table 2. Pit 9 Radiological Inventory in 1991 and 1992

| Isotope | 1991 Radio-activity (Ci) | 1991 Mass (g) | 1992 Radio-activity (Ci) | 1992 Mass (g) |
|---|---|---|---|---|
| U-234 | 8.23E-02 | 1.32E+01 | 8.23E-02 | 1.32E+01 |
| U-235 | 3.75E-03 | 1.73E+03 | 3.75E-03 | 1.73E+03 |
| U-238 | 3.97E+00 | 1.18E+07 | 3.97E+00 | 1.18E+07 |
| Th-234 | 3.97E+00 | 1.72E-04 | 3.97E+00 | 1.72E-04 |
| Pu-238 | 2.57E+01 | 1.50E+00 | 2.55E+01 | 1.49E+00 |
| Pu-239 | 1.16E+03 | 1.87E+04 | 1.16E+03 | 1.87E+04 |
| Pu-240 | 2.65E+02 | 1.17E+03 | 2.65E+02 | 1.17E+03 |
| Pu-241 | 3.07E+03 | 2.97E+01 | 2.93E+03 | 2.84E+01 |
| Pu-242 | 1.26E-02 | 3.20E+00 | 1.26E-02 | 3.20E+00 |
| Am-241 | 2.26E+03 | 6.59E+02 | 2.26E+03 | 6.59E+02 |
| Co-60 | 1.46E-02 | 1.29E-05 | 1.28E-02 | 1.13E-05 |
| Co-60(MAP) | 5.83E-04 | 5.16E-07 | 5.11E-04 | 4.52E-07 |
| Cs-137(MFP) | 2.63E+00 | 3.04E-02 | 2.57E+00 | 2.97E-02 |
| Ba-137m(MFP) | 2.49E+00 | 4.63E-09 | 2.43E+00 | 4.52E-09 |
| Sr-90(MFP) | 2.38E+00 | 1.71E-02 | 2.33E+00 | 1.68E-02 |
| Y-90(MFP) | 2.38E+00 | 4.38E-06 | 2.33E+00 | 4.29E-06 |

Table 2 displays the expected radioactivity in Curies and mass in grams for 1991 and 1992. The decay corrections were conducted using RADDECAY 2.05[11] and/or PDACT 1.1[12]. Both programs employ the Bateman equations but RADDECAY is limited in that it cannot simultaneously incorporate a nonzero daughter activity and it also has fixed half-lives and branching ratios. PDACT allows the user to specify the decay parameters of choice and allows a nonzero daughter activity. It is limited to parent-daughter decay and does not proceed further down a decay chain.

Table 2 includes 3 daughters that are not shown in Table 1. These are Th-234 (daughter of U-238), Ba-137m, and Y-90 with half-lives of 6.598E-2 y, 4.852E-6 y and 7.310E-3 y, respectively (see Reference 8). The calculations have been limited to the significant daughters of specific radionuclei. No quantities of granddaughter radionuclei are included.

The inventories in Tables 1 and 2 represent estimates of radionuclides that have been derived from Rocky Flats data, shipping records and RWMIS data. The Rocky Flats values are limited in that we are unable to substantiate the uncertainty associated with the 1968 quantities of each radioisotope stated by Lee (see Reference 1). The fact that we have assumed complete mixing of these radioisotopes for selected volumes of 1968 Rocky Flats waste delivered to the INEL may be in error, but it represents a reasonable option without any other information. The onsite MAP and MFP wastes are represented Co-60, and Cs-137 and Sr-90 with their daughters, respectively. These choices are appropriate for transient and shorter irradiation times (times smaller than the half-lives). The predominant radionuclides after 100 or more years may change for the MFP's, but this is dependent on the irradiation history for a specific waste generator source. The contributon to risk after 100 years from MAP and MFP in Pit 9 is quite small relative to the expected risk arising from the uranium and transuranic waste.

## References


1. Letter from W. H. Lee to H. F. Soule, "Rocky Flats Solid Waste Shipped To NRTS", (received at the INEL December, 1971).

2. Draft Letter form D. W. Rhodes to J. L. Butler, "Amounts of Pu-239 disposed in Pit 9 in 1968", May 30, 1991.

3. E. C. Garcia and J. L. Knight, "Detailed Estimate of Radioactive Material Contents for Pit 9", Engineering Design File BWP-ISV-004 Rev.0, February 23, 1989.

4. J. K. Hartwell and D. N. Thompson, *Investigation of a Gamma-Ray Spectrometric Low Level Waste Measurement System: FY 1988 Activities*, EG+G Internal Technical Report ST-CS-028-88, September 1988.

5. R. L. Bunting, *INEL (Data Master) Isotope Library*, January 1983 (Unpublished).

6. B. F. Rider, *Compilation of Fission Product Yields, Vallecitos Nuclear Center, 1981*, NEDO-12154-3(C), ENDF-322, Class 1, October 31, 1981.

7. General Electric Co., *Chart of the Nuclides (Thirteenth Edition)*, 1984.

8. J. K. Tuli, *Nuclear Wallet Cards*, National Nuclear Data Center, Brookhaven National Laboratory, January 1985.

9. C. M. Lederer and V. S. Shirley, Editors, *Table of Isotopes (7th Edition)*, Lawrence Berkeley Laboratory, John Wiley & Sons Inc., 1978.

10. K. Z. Morgan and J. E. Turner, Editors, *Principles of Radiation Protection: A Textbook of Health Physics*, John Wiley & Sons, Inc., 1967.

11. Grove Engineering, *RADDECAY Version 2.05*, October 1987.

12. J. J. King, *PDACT Version 1.1*, (Written with Quick Basic 4.5 using double precision), July 1, 1991.